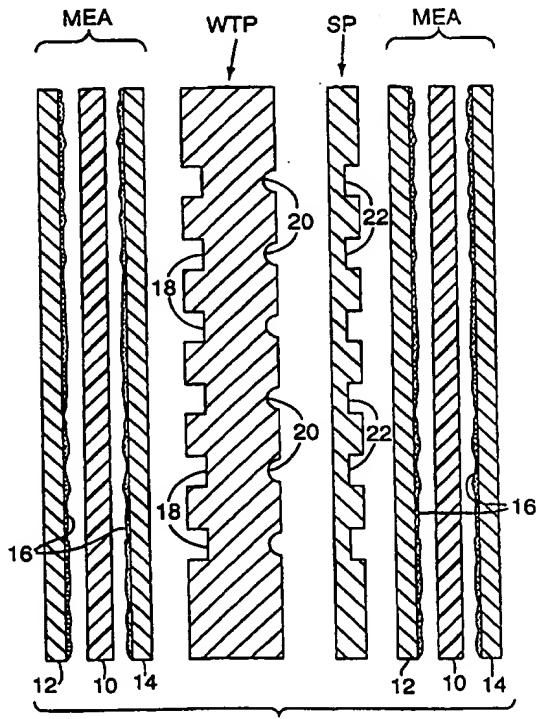


PCTWORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁷ : H01M 2/08, 2/14	A1	(11) International Publication Number: WO 00/24066 (43) International Publication Date: 27 April 2000 (27.04.00)
(21) International Application Number: PCT/US99/24629 (22) International Filing Date: 21 October 1999 (21.10.99) (30) Priority Data: 09/176,355 21 October 1998 (21.10.98) US (71) Applicant: INTERNATIONAL FUEL CELLS, LLC [US/US]; 195 Governor's Highway, South Windsor, CT 06074 (US). (72) Inventors: KRASIJ, Myron; 181 Deercliff Road, Avon, CT 06001 (US). MOSKEY, Edward, A.; 38 Coe Avenue, Portland, CT 06480 (US). (74) Agent: KELLY, Robert, H.; Hamilton Sundstrand, One Hamil- ton Road, Windsor Locks, CT 06074 (US).		(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>
(54) Title: FUEL CELL WITH IMPROVED SEALING BETWEEN INDIVIDUAL MEMBRANE ASSEMBLIES AND PLATE ASSEMBLIES		
(57) Abstract <p>A fuel cell stack includes a plurality of fuel cells, each of which includes a membrane electrode assembly (MEA) and a water transport plate (WTP), or a fluid flow plate fabricated from graphite. This plate and optionally a separator plate (SP) are held in assembled relationship with one another and with the membrane electrode assemblies (MEA) by a fluoroelastomeric adhesive/sealant (30) that is also coated on the external edges of these components to provide a water-tight seal to better contain the coolant fluid in the form of water provided in the fuel cell stack.</p> 		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav	TM	Turkmenistan
BF	Burkina Faso	GR	Greece		Republic of Macedonia	TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's	NZ	New Zealand		
CM	Cameroon		Republic of Korea	PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

DESCRIPTIONFUEL CELL WITH IMPROVED SEALING BETWEEN INDIVIDUAL
MEMBRANE ASSEMBLIES AND PLATE ASSEMBLIESTECHNICAL FIELD

- 5 This invention relates to electrochemical fuel cells, and deals more particularly with the sealing provided between a coolant flow-field plate and an adjacent component of the fuel cell, particularly with the membrane electrode assembly and/or an associated water separator plate.

BACKGROUND ART

- 10 Prior patents 5, 264,299 and 5,523,175 assigned to the assignee herein are representative of a prior art approach to sealing, with an elastomeric sealing material, peripheral portions of fuel cell components, particularly components which may be porous. More particularly, column 4 line 1 through column 6 line 49 of the '299 patent describes how a support
15 plate might be assembled with a proton exchange membrane by such a sealing material peripherally joining these components. The '299 patent is hereby incorporated by reference herein.

- Peripheral sealing of prior art membrane and plate assemblies generally requires impregnating the outside perimeter of each component
20 with a chemical, such as SARTOMER GRADE 2108 from SARTOMER CO., INC., that requires curing in the absence of oxygen. Such chemicals are hydrophobic in the cured state so as to suffer from continuous contact with the cooling fluid currently favored, namely water as now used in fuel cells generally.

- 25 In prior art fuel cells the edges of water transport plates are impregnated with a resin, which when polymerized create an edge seal that prevents leakage of the gaseous reactants, and also serves to prevent infiltration of these reactants into the liquid coolant.

- Typically resins that have been used to create such edge seals in
30 water transport plates are SARTOMER 2100 made by SARTOMER CO.,

INC., and SYLGARD 170 made by the Dow Chemical Corporation. These polymers are somewhat hydrophobic, however, and are not totally wetted by water. Since water is a product of the reaction within the fuel cell, and typically also comprises the coolant in the proton exchange membrane fuel cell (PEM) such characteristics are undesirable for use in such edge seal applications. More particularly, as a consequence of this hydrophobicity is that the interface between the central active area of the water transport plate and the peripheral edge seal portion of the water transport plate may not totally fill with water. Such a situation reduces the sealing effectiveness of the water transport plate, and results in some leakage of reactant gases into the coolant which can result in a undesirable condition.

The edges of the porous catalyst support plates are impregnated to create edge seals also. One commonly used compound for such impregnation is a silicone rubber. However, the silicone rubber although resulting in an effective seal within the catalyst support plate does not exhibit good adhesion with the proton exchange membrane (PEM). This poor adhesion tends to permit reactant leakage at the interface between these components and allows the PEM to shrink away from the edge of the catalyst support plate, particularly if the water content of the proton exchange membrane is reduced due to the conditions of the PEM operating fuel cell. The shrinkage of the proton exchange membrane can result in an electrical short between the anode catalyst support plate and the cathode catalyst support plate. Such a short will of course reduce the efficiency of the fuel cell and may cause localized overheating.

Finally, the seal configuration in a typical PEM cell generally employs use of an acrylic tape to bond the periphery of the water transport plates to the periphery of the catalyst support plates in order to create an effective seal between these components. Such acrylic tape is also used to bond the two halves of the water transport plate to each other. Such seal configurations exhibit marginal performance due to hydrophobicity of the materials that are used and due to poor adhesion with the PEM. Further, such seals are expensive to manufacture as a result of the number of components and process steps required in their fabrication and assembly.

One object of the present invention is to provide a single peripheral sealant coating that adheres to the porous graphite water transport plates and the porous graphite catalyst support plates, and which also adheres to the exchange membrane itself.

- 5 An advantage of the present invention can be attributed to the fact that this single adhesive/sealant that is used to coat the exterior edges and to impregnate the exterior edges of these components provides a convenient way of achieving desired functions of both sealing and of adhesion, and also obviates the disadvantages created by use of the materials described above for achieving the sealing and adhesion functions.
- 10

DISCLOSURE OF INVENTION

- The details of the fuel cell construction and its method of operation are well known and described in commonly owned U.S. patent number 5,264,299 and U.S. patent number 5,523,175 as well as U.S. patent number 15 5,303, 944 and U.S. patent number 5,700,595 plus 4,769,297 all of which patents are incorporated by reference herein.

- These and other objects of the present invention can be achieved by providing a fuel cell stack wherein the individual cells in the stack include a membrane electrode assembly (MEA) wherein opposed catalyst layers are 20 provided on opposite sides of the proton exchange membrane (PEM), and wherein opposed anode and cathode support plates, typically of porous graphite, are provided on opposite sides of this catalyst, and wherein said catalyst support plates contain a central area adjacent to the catalyst and peripheral areas to be sealed including a first seating surface. The combined reactant flow field plate/water transport plate is provided adjacent 25 to the catalyst support plate and is also of porous graphite and includes a peripheral sealing area including a second seating surface.

- A sealant coating preferably in the form of a fluoroelastomer is provided between these first and second seating surfaces, and serves to 30 adhere and to bond the catalyst support plate to the flow-field plate.

The catalyst support plate provided adjacent to the catalyst layer and the membrane itself presents some problem in prior art fuel cell

configurations, but the same sealant coating material can also be provided around the edges of the membrane electrode assembly and associated water transport plate or plates to provide a structure that prevents leakage of reactants and coolant.

5 BRIEF DESCRIPTION OF THE DRAWINGS

These and other features, aspects, and advantages of the present invention will become better understood with regard to the following description, appended claims, and accompanying drawings where:

FIG. 1 is an exploded side elevational view illustrating several of the
10 components in a pair of side-by-side fuel cells in a typical fuel cell stack of the type adapted to take advantage of the present invention;

FIG. 2 is a view of a portion of the fuel cell of Fig. 1 showing the sealant coating provided between peripheral surfaces of the indicated components of the fuel cell, and also showing the same sealant coating
15 provided at the outer edge of the fuel cell stack.

FIG. 3 is a view similar to FIG 2 but showing the effect of the porous catalyst support plate and water transport plate assemblies for a cell with two water transport plates.

BEST MODE FOR CARRYING OUT THE INVENTION

20 Turning now to the drawings in greater detail, Fig. 1 shows, in exploded relationship, the various components of a typical fuel cell which are provided in repeating fashion in a typical fuel cell stack. Two membrane electrode assemblies are indicated at **MEA** in Fig. 1, and interposed therebetween is provided a water transport plate **WTP** and a separator plate
25 **SP**.

Each membrane electrode assembly comprises a solid polymer ion exchange membrane **10** interposed between an anode **12** and a cathode **14**. The anode and cathode will be referred to hereinafter as electrodes, and each contains a catalyst layer, such as indicated generally at **16** in Fig.
30 **1** and a catalyst support plate. The catalyst layer is provided on the side of the electrode adjacent to the membrane **10**, and renders these electrodes

electrochemically active.

A flow-field plate, or water transport plate **WTP** contains channels **18** on the side facing the membrane electrode assembly **MEA** through which channels **18** an oxidant usually in the form of oxygen or air flows. This flow-field plate **WTP** contains other channels **20** on the side facing away from the membrane electrode assembly **MEA** through which channels **20** a coolant fluid in the form of water flows for purposes of withdrawing heat from the electrochemical reaction provided in the fuel cell.

A separator plate **SP** is provided adjacent to the water transport plate **WTP** for closing these channels and to define, on its opposite side adjacent the next membrane electrode assembly, suitable channels, such as shown at **22**, for the flow of fuel, usually in the form of a gas containing hydrogen, adjacent the anode **12**.

Variations of this structure are possible, and in some installations it may be possible or preferable to define the channels **20** for the coolant in a separator plate **SP** rather than in the same plate which defines the channels **18** associated with a reactant gas.

The prior art suggests that the seal between these various components, namely the membrane electrode assembly, the water transport plate, and the separator plate **SP**, comprise interfacial peripheral gaskets of silicone coated glass fabric preferably in the form of a silicone pressure sensitive adhesive applied to one side of each of these components. These silicone gaskets do provide acceptable sealing, but require relatively high sealing loads, and must, of necessity, be cut out of large sheets, creating a great deal of waste. Other approaches to this problem have taken the form of an edge seal formed by impregnating the outside perimeter of these components with a resin, such as SARTOMER. Unfortunately this resin produces marginal results because such resins are typically hydrophobic in the cured state and therefore suffer from a reduction in the water bubble pressure at the edge seal interface. After impregnation with water, poor wetting of the chemical coated components generally occurs resulting in local leakage.

As mentioned previously, the preferred form of electrode support

plate is generally in sheet form, or paper form, and is coated with the catalyst and requires sealing along its peripheral edges with a silicone rubber, such as SYLGARD 170. Use of such a silicone sealant provides an acceptable seal, but does not afford sufficient adhesion with the membrane electrode assembly itself, which is typically fabricated from NAFION.

In accordance with the present invention, an improved adhesive/sealant 30 is provided both on the outer edges of the cell components and between the membrane electrode assembly and the water transport plate, as best shown in Fig. 2. The preferred material for such sealant is a fluoroelastomer, such as FLUOROLAST grade SB or WB manufactured by Fluorolast, a division of Laureen. Not only does this brand of fluoroelastomer have excellent adhesion with NAFION, as used in connection with construction with the membrane electrode assembly MEA, but the FLUOROLAST has also been found to adhere very well to graphite. The graphite material currently comprising the preferred conductive material for manufacturing fluid-flow plates, such as those illustrated at WTP and SP in Fig. 2. Graphite is suited for use with such an adhesive/coating so that these components will be secured to one another and afford an effective seal against the water in the channels 20.

Alternatively the separator plate may be eliminated and two water transfer plates (WTP) may be used in a back to back configuration. In this configuration the reactant flow channels are defined on the outward surfaces, and the water flow channels are defined on the inward surfaces. Each WTP is fabricated with a sufficient bubble pressure that it provides the function of separating the fuel from the oxidant.

The preferred embodiment of the invention is shown in FIG. 3 where the periphery of the electrode support plate is impregnated with the Fluorolast elastomer. FIG. 3 also shows the alternative cell configuration with back to back water transport plates (WTP) that eliminates the separator plate. The periphery of the electrode support plate is pervious, and impregnated with the Fluorolast elastomer to create an edge seal and the sealing surface of each WTP is coated with Fluorolast.

Laboratory results show that the variety of interfacial seals and edge

seals formerly required, in the form of acrylic tape, SARTOMER 2100 and SYLGARD 170 can be replaced with the fluoroelastomer material 30 shown in figure 2 thereby reducing the overall cost of manufacture of a typical fuel cell stack and improving the sealing characteristics.

5 Subscale component tests have been performed to verify the various features of the invention. More particularly a 14 mil porous graphite electrode support plate, grade TGP-H-120 sold by Toray, was impregnated with Flourolast, and following impregnation the sample was cured for about 1 hour at about 212°F. The sample was then cut into a 3 inch by 3 inch
10 frame, and so as to have a frame width of 0.25 inches. Two such samples were constrained in a pressure test fixture, and no overboard leakage of nitrogen was measured when the interior of the frame was pressurized to 95 psig (gauge pressure in pounds per square inch).

 A similar test was performed where the sealing surface of a,
15 separator plate being of dense graphite material, and 0.9mm thick, of the type sold by KOBE STEEL LTD. under its grade number GCR 101 G. These components were coated with Flourolast and cured for 72 hours at room temperature of approximately 70°F. Four such samples were cut to the above dimensions and assembled into a pressure test fixture. The
20 bonded stack withstood 95 psig internal pressure of nitrogen with no inter facial leakage as evidence by the absence of a pressure decay for ten minutes. No gas bubbles were seen to form when the stack was immersed in water.

 Tests were also performed to evaluated the effect of the
25 flouroelastomer edge seal on the bubble pressure of the water transport plates. 3 inch by 3 inch by 0.1 inch thick porous graphite water transport plates made by International Fuel Cells were filled with water and their bubble pressures determined to be 8 psi. The samples were then dried, their edges sealed with the flourolast, and 3 inch by 3 inch by 2.5 graphite
30 frames bonded to both sides of the sample with Flourolast. At the curing of 180 °F for 16 hours the samples were again filled with water and the bubble pressure measured to be 8 psi, indicating that edge sealing with the flouroelastomer did not degrade the bubble pressure of the water transport

curing of 180 °F for 16 hours the samples were again filled with water and the bubble pressure measured to be 8 psi, indicating that edge sealing with the flouroelastomer did not degrade the bubble pressure of the water transport plates. This particular sample was then tested for outboard leaks
5 as discussed above with no leakage measured at 95 psi.

While preferred embodiments have been shown and described, various modifications and substitutions may be made without departing from the spirit and scope of the invention. Accordingly, it is to be understood that the present invention has been described by way of example, and not by
10 limitation.

CLAIMS

1. A fuel cell stack for generating electrical energy, wherein the cells in the stack are comprised of:
- a membrane electrode assembly including opposed catalyst layers, and a proton exchange membrane between said catalyst layers,
 - 5 including opposed anode and cathode support plates adjacent to the catalyst layers, each catalyst support plate having a marginal peripheral face defining a first sealing surface;
 - a reactant flow-field plate or water transport plate adjacent to one said catalyst support plate and having a peripheral face defining a
 - 10 second sealing surface; and
 - a sealant coating between first and second sealing surfaces, said sealant coating being an elastomeric adhesive for bonding said catalyst support plate to said flow-field plate or water transport plate.
2. The combination according to Claim 1, wherein said membrane electrode assembly includes a catalyst support layer between said catalyst layer and said membrane, said catalyst support layer provided in the form of a support sheet, and said sealant coating also provided
- 5 externally around said membrane electrode assembly and also externally around said reactant flow-field plate.
3. The combination according to Claim 1, wherein said elastomeric adhesive comprises a fluoroelastomer.
4. The combination according to claim 1, wherein said elastomeric adhesive is Fluorolast.

5. The combination according to Claim 1, further characterized by a separator plate provided adjacent said flow-field plate to define coolant flow passageways therebetween, and both said flow-field plate and said separator plate being sealed in the areas of their peripheries by said elastomeric adhesive.

6. The combination according to Claim 1, wherein said fluid flow-field plate is fabricated from a graphite material.

7. The combination according to Claim 1, wherein said catalyst support plates are fabricated from a graphite material.

8. The combination according to any of the preceding claims, wherein said sealant coating comprises a fluoroelastomer, and wherein said fluoroelastomer is provided externally of said membrane electrode assembly and flow-field plates or water transport plates in a fuel cell stack.

9. The combination according to claim 1 wherein said catalyst support plates have pervious peripheral portions impregnated with a fluoroelastomer, and wherein said elastomeric adhesive comprises a fluoroelastomer.

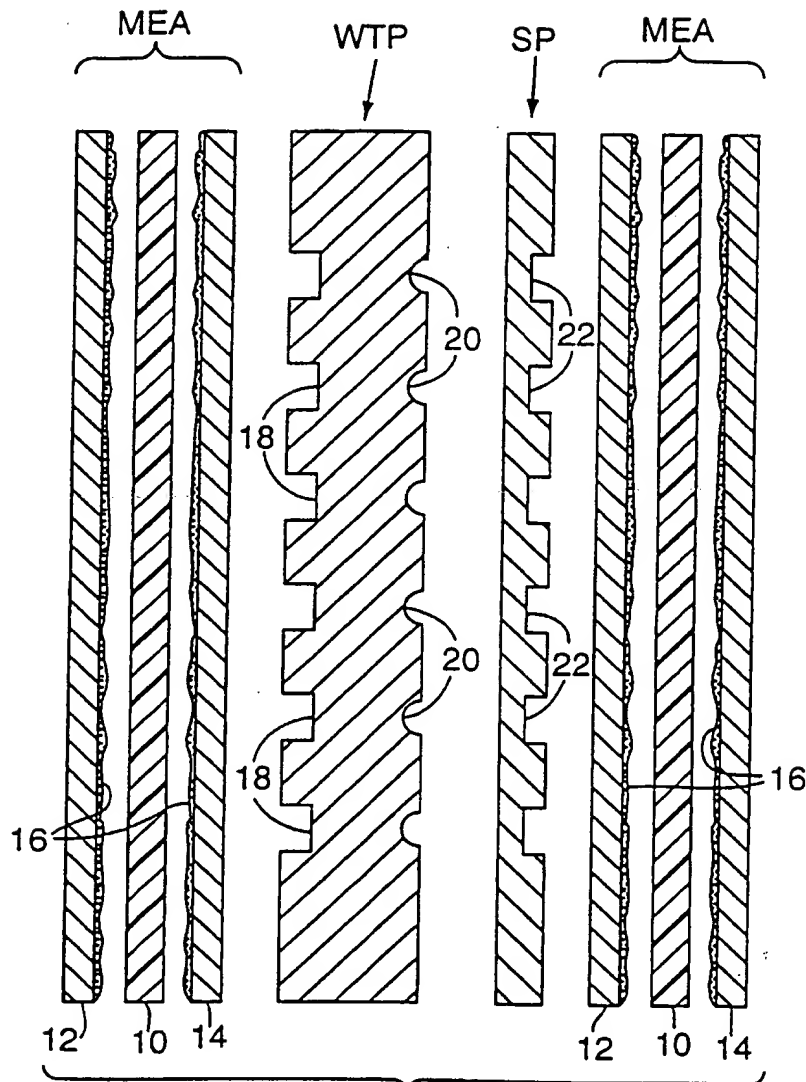


FIG. 1

2/2

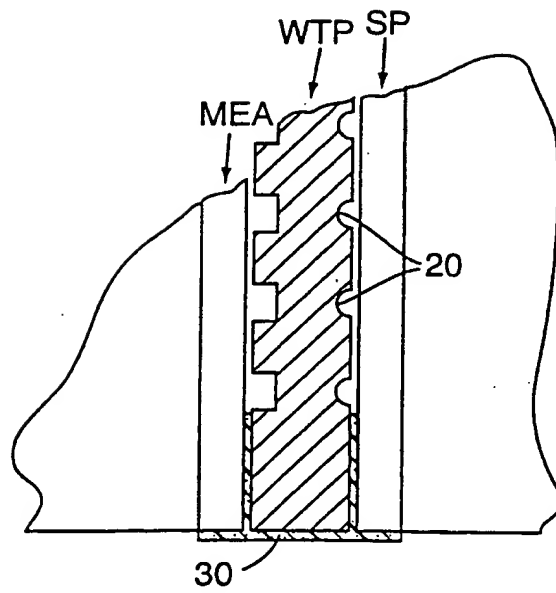


FIG. 2

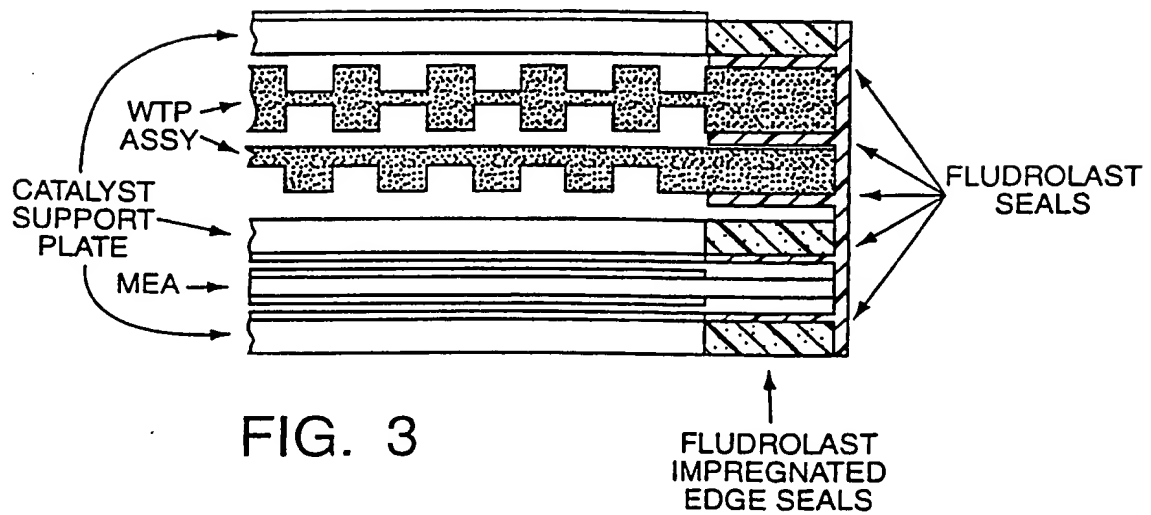


FIG. 3

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/24629**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(7) : H01M 2/08, 2/14

US CL : 429/34-36, 38, 39

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 429/34-36, 38, 39

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5,328,542 A (GRASSO ET AL) 12 July 1994, col. 4, lines 1-59; col. 6, lines 32-60.	1-9
X	US 4,774,154 A (SINGELYN ET AL) 27 September 1988, col. 2, lines 22-67 and col. 4, lines 6-31.	1-4, 8, 9
A	US 4,374,185 A (POWERS ET AL) 15 February 1983.	1-4, 8, 9

☐ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

* Special categories of cited documents:	*T	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be of particular relevance	*X*	document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
E earlier document published on or after the international filing date	*Y*	document of particular relevance, the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*Z*	document member of the same patent family
O document referring to an oral disclosure, use, exhibition or other means		
P document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search 10 JANUARY 2000	Date of mailing of the international search report 07 FEB 2000
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer TRACY DOVE Telephone No. (703) 305-0661